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71 Applicant: TDK Corporation
13-1, Nihonbashi 1-chome Chuo-Ku
Tokyo-to(JP)

72 Inventor: Ono, Shuichi
74, Shinkawa Aza Miyonoshita
Honjou-shi Akita-ken(JP)
Inventor: Yahagi, Masahiro
200 Ryouzenji Aza Kitsunemori
Nikahomachi Yuri-gun Akita-ken(JP)
Inventor: Itagaki, Shuichi
23-1, Yachimachi, Honjou-shi
Akita-ken(JP)
Inventor: Kikuchi, Nobuaki
1-28, Torinoumi Kusakatamachi
Yuri-gun Akita-ken(JP)

74 Representative: Eisenführ & Speiser
Martinistrasse 24
D-2800 Bremen 1(DE)

54 Semiconductive ceramic composition.

57 A semiconductive ceramic composition for a reduction reoxidation type semiconductive ceramic capacitor capable of increasing capacitor, dielectric strength of the capacitor and improving temperature characteristics of the capacitor. The composition includes a BaTiO₃ base component and a minor component consisting of Nb and Ce. Nb and Ce are present in amounts of 0.2 to 3.0 mol% on Nb₂O₅ and CeO₂ bases. Addition of Co and the like increases insulation resistance and D.C. breakdown voltage of the capacitor and further improves temperature characteristics of the capacitor.

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SEMICONDUCTIVE CERAMIC COMPOSITION

BACKGROUND OF THE INVENTIONField of the Invention

This invention relates to a semiconductive ceramic composition for a semiconductive ceramic capacitor, and more particularly to a semiconductive ceramic composition suitable for use for manufacturing of a reduction and reoxidation type semiconductive ceramic capacitor.

Description of the Prior Art

A semiconductive ceramic capacitor serving as a passive electronic circuit element is generally classified into two types or a boundary-layer type and a surface-layer type. The boundary-layer type semiconductive ceramic capacitor includes a boundary insulation type one and the surface-layer type semiconductive ceramic capacitor includes a barrier-layer type one and a reduction reoxidation type one.

The semiconductive ceramic capacitor, particularly, reduction reoxidation type semiconductive ceramic capacitor generally has a disadvantage that an increase in unit area capacity expressed at $\mu\text{F}/\text{cm}^2$ causes a significant decrease in breakdown voltage, resulting in an increase in dielectric loss and/or deterioration of temperature characteristics.

SUMMARY OF THE INVENTION

The present invention has been made in view of the foregoing disadvantage of the prior art.

Accordingly, it is an object of the present invention to provide a semiconductive ceramic composition for a semiconductive ceramic capacitor which is capable of causing the capacitor to exhibit not only increased unit area capacity but increased dielectric strength and excellent temperature characteristics.

It is another object of the present invention to provide a semiconductive ceramic composition for a semiconductive ceramic capacitor which is capable of improving insulation resistance and D.C. breakdown voltage of the capacitor.

It is a further object of the present invention to provide a semiconductive ceramic composition for a reduction reoxidation type semiconductive ceramic capacitor which is capable of causing the capacitor to exhibit not only increased unit area capacity but increased dielectric strength and excellent temperature characteristics.

It is still another object of the present invention to provide a semiconductive ceramic composition for a reduction reoxidation type semiconductive ceramic capacitor which is capable of improving insulation resistance and D.C. breakdown voltage of the capacitor.

In accordance with the present invention, a semiconductive ceramic composition is provided. The composition comprises a BaTiO_3 base component and a minor component comprising Nb and Ce. Nb and Ce are present at 0.2 to 3.0 mol% on Nb_2O_5 and CeO_2 bases, respectively.

In accordance with the present invention, a semiconductive ceramic composition is also provided which comprises a BaTiO_3 base component, a minor component comprising Nb and Ce, and an additive component comprising Co, Mn, SiO_2 and SrTiO_3 . Nb and Ce are present at 0.2 to 3.0 mol% on Nb_2O_5 and CeO_2 bases, respectively. Co and Mn are present at 0.8 wt% or less and 0.25 wt% or less on Co_3O_4 and MnCO_3 bases, respectively. SiO_2 and SrTiO_3 are present in amounts of 0.25 wt% or less and 0.5 to 20.0 wt%. The additive component may comprise Co, Mn, SiO_2 and CaTiO_3 , wherein Co, Mn and SiO_2 may be present in the same amounts as described above. CaTiO_3 may be present at 0.2 to 15.0 wt%. Alternatively, the additive component may comprise Co, Mn, SiO_2 and Y. Co, Mn and SiO_2 may be present in the same amounts as described above. Y may be present at 0.1 to 3.0 wt% on a Y_2O_3 basis.

DETAILED DESCRIPTION OF THE INVENTION

The present invention is directed to a semiconductive ceramic composition for a semiconductive ceramic capacitor, particularly, a reduction reoxidation type semiconductive ceramic capacitor which comprises a BaTiO_3 base component and a minor component comprising Nb and Ce. The term "minor component" used herein indicates an essential component smaller in quantity than the base component. Nb and Ce constituting the minor component are present in amounts of 0.2 to 3.0 wt% based on Nb_2O_5 and CeO_2 bases, respectively. It was found that use of the composition of such construction for a semiconductive ceramic capaci-

tor, particularly, a reduction and reoxidation type one causes the capacitor to be significantly increased in capacity and dielectric strength and effectively exhibit good temperature characteristics. The composition may also contain Co in an amount of 0.8 wt% or less on a Co_3O_4 basis. Such addition of Co significantly improves insulation resistance and D.C. breakdown voltage of the capacitor and causes it to exhibit more temperature characteristics. Further, the composition may contain Mn in an amount of 0.25 wt% or less on a MnCO_3 basis. In addition, it may contain SiO_2 in an amount of 0.25 wt% or less. Incorporation of Mn and SiO_2 further promotes the above-noted advantage obtained due to addition of Co.

The present invention is also directed to a semiconductive ceramic composition for a semiconductive ceramic capacitor, particularly, a reduction reoxidation type semiconductive ceramic capacitor which comprises a BaTiO_3 base component, a minor component comprising Nb and Ce, and an additive component comprising Co, Mn, SiO_2 and SrTiO_3 . Nb and Ce are present in amounts of 0.2 to 3.0 mol% on Nb_2O_5 and CeO_2 bases, respectively. Co and Mn are present in amounts of 0.8 wt% or less and 0.25 wt% or less on Co_3O_4 and MnCO_3 bases, respectively. SiO_2 and SrTiO_3 are present in amounts of 0.25 wt% or less and 0.5 to 20.0 wt%, respectively.

Use of the so-formulated composition for a semiconductive ceramic capacitor, particularly, a reduction and reoxidation type one likewise causes the capacitor to be substantially increased in capacity and dielectric strength and positively exhibit good temperature characteristics.

The composition may further contain Y, which may be present in an amount of 0.1 to 3.0 wt% on a Y_2O_3 basis. Addition of Y causes the capacitor to exhibit improved insulation resistance and D.C. breakdown voltage and more temperature characteristics.

The present invention is further directed to a semiconductive ceramic composition for a semiconductive ceramic capacitor, particularly, a reduction reoxidation type semiconductive ceramic capacitor which comprises a BaTiO_3 base component, a minor component comprising Nb and Ce, and an additive component comprising Co, Mn, SiO_2 and CaTiO_3 . Nb and Ce are present in amounts of 0.2 to 3.0 mol% on Nb_2O_5 and CeO_2 bases, respectively. Co and Mn are present in amounts of 0.8 wt% or less and 0.25 wt% or less on Co_3O_4 and MnCO_3 bases, respectively. SiO_2 and CaTiO_3 are present in amounts of 0.25 wt% or less and 0.2 to 15.0 wt%, respectively.

Use of the so-formulated composition for a semiconductive ceramic capacitor, particularly, a reduction and reoxidation type one likewise causes the capacitor to be increased in capacity and dielectric strength and exhibit good temperature characteristics.

The composition may further contain Y, which may be present in an amount of 0.1 to 3.0 wt% on a Y_2O_3 basis. Addition of Y causes the capacitor to exhibit improved insulation resistance and D.C. breakdown voltage and more temperature characteristics.

Furthermore, the present invention is directed to a semiconductive ceramic composition for a semiconductive ceramic capacitor, particularly, a reduction reoxidation type semiconductive ceramic capacitor which comprises a BaTiO_3 base component, a minor component comprising Nb and Ce, and an additive component comprising Co, Mn, SiO_2 and Y. Nb and Ce are present in amounts of 0.2 to 3.0 mol% on Nb_2O_5 and CeO_2 bases, respectively. Co and Mn are present in amounts of 0.8 wt% or less and 0.25 wt% or less on Co_3O_4 and MnCO_3 bases, respectively. SiO_2 is present in an amount of 0.25 wt% or less and Y is present in an amount of 0.1 to 3.0 wt% on a Y_2O_3 basis. Use of the so-formulated composition for a semiconductive ceramic capacitor, particularly, a reduction and reoxidation type one likewise causes the capacitor to be increased in capacity and dielectric strength and exhibit improved insulation resistance and D.C. breakdown voltage and good temperature characteristics.

Each of the so-formulated semiconductive ceramic composition may be compacted into a semiconductive ceramic element and sintered to cause a surface layer of the element to be insulated.

The invention will be understood more readily with reference to the following examples, however, these examples are intended to illustrate the invention and are not to be construed to limit the scope of the invention.

Example 1

BaCO_3 , TiO_2 , Nb_2O_5 , CeO_2 , Co_3O_4 , MnCO_3 and SiO_2 each having an industrial grade were used as starting materials. The materials were weighed so that each composition may be obtained which has a composition ratio as shown in Table 1. The materials were subjected to wet blending in a ball mill of synthetic resin using water and pebbles for 20 hours while stirring to prepare a composition mixture. Then, the so-obtained mixture was de-watered and dried, and provisionally burned at 1200°C for 2 hours. Subsequently, the mixture was powdered and blended for 20 hours by means of

the ball mill and then dewatered and dried, to which polyvinyl alcohol (PVA) in an amount of 2 wt% was added as an organic binder to carry out granulation and grading to prepare granulated powder of the composition. The powder was then formed into a disc-like compact or semiconductive ceramic element of 10mm in diameter and 0.5mm in thickness at compacting pressure of about 3 tons/cm². The element was heat-treated at 800°C for 1 hour to remove the binder therefrom and then subjected to burning at about 1300°C for about 2 hours. Then, it was subjected to reduction at 800°C for 2 hours in a stream of a reducing atmosphere (H₂ atmosphere) to prepare a reduction reoxidation type semiconductive ceramic element, which was then heat-treated at 800°C for 2 hours in an oxidizing atmosphere (air). This caused reoxidation of only a surface of the element, resulting in the surface being provided with an insulating property. Thereafter, an Ag paste was applied onto both surfaces of the semiconductive ceramic element by printing and then baked at 800°C to form electrodes, resulting in a reduction reoxidation type semiconductive ceramic capacitor.

A test was then carried out to consider electrical characteristics of the so-manufactured capacitor. The results were as shown in Table 1, wherein capacity and dielectric loss ($\tan \delta$) were measured at a frequency of 1kHz and insulation resistance was measured at a room temperature of 20°C while applying D.C. voltage of 25V to the capacitor.

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Table 1

*1	Specimen	Composition Ratio						Electrical Characteristics					
		mol%			wt%			C ^{*2} ($\mu\text{F}/\text{cm}^2$)	tan δ ^{*3} (%)	IR ^{*4} (M Ω) (10 ³)	Vb ^{*5} (V)	T.C. ^{*6} (%)	
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃	SiO ₂					-25°C	+85°C
X	1	99.8	0.1	0.1	0	0	0	0.02	10.0	0.5	700	0	-10
X	2	99.4	0.1	0.5	0	0	0	0.05	7.0	0.7	500	+1	-13
X	3	95.9	0.1	4.0	0	0	0	0.10	8.0	0.7	500	+8	-30
	4	99.6	0.2	0.2	0	0	0	0.15	4.3	5	800	+3	-10
	5	99.3	0.2	0.5	0	0	0	0.18	4.0	5	700	+3	-15
	6	96.8	0.2	3.0	0	0	0	0.25	4.3	7	1000	-1	-27
	7	99.0	0.5	0.5	0	0	0	0.27	3.5	5	700	+4	-18
	8	98.0	0.5	1.5	0	0	0	0.30	3.7	5	900	+4	-23
	9	98.5	1.0	0.5	0	0	0	0.23	3.5	5	700	+4	-19
X	10	98.4	1.5	0.1	0	0	0	0.05	11.0	0.7	500	+5	-13
	11	98.3	1.5	0.2	0	0	0	0.17	4.3	5	700	+5	-15
	12	98.0	1.5	0.5	0	0	0	0.20	4.0	5	700	+5	-20
	13	97.5	1.5	1.0	0	0	0	0.21	3.5	5	800	+3	-23

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Table 1 (Continued)

#1	Specimen No.	Composition Ratio						Electrical Characteristics						
		mol%			wt%			C*2 ($\mu\text{F}/\text{cm}^2$)	$\tan\delta$ *3 (%)	IR*4 (M Ω) (10^3)	Vb*5 (V)	T.C.*6 (%)		
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃	SiO ₂					-25°C	+85°C	
		14	97.0	1.5	1.5	0	0	0	0.23	3.7	5	700	+1	-25
		15	96.5	1.5	2.0	0	0	0	0.25	3.8	5	800	0	-28
		16	95.5	1.5	3.0	0	0	0	0.26	4.0	5	1000	-3	-30
X		17	94.5	1.5	4.0	0	0	0	0.28	7.0	5	1000	-10	-40
		18	97.5	2.0	0.5	0	0	0	0.20	4.0	5	700	+3	-22
		19	96.5	2.0	1.5	0	0	0	0.23	4.0	5	800	+4	-26
		20	96.8	3.0	0.2	0	0	0	0.15	4.0	5	700	+5	-20
		21	96.5	3.0	0.5	0	0	0	0.15	4.5	5	500	+5	-23
		22	94.0	3.0	3.0	0	0	0	0.20	4.3	5	800	-2	-30
X		23	95.9	4.0	0.1	0	0	0	0.05	12.0	0.5	300	+13	-25
X		24	95.5	4.0	0.5	0	0	0	0.07	8.0	0.7	300	+7	-30
X		25	92.0	4.0	4.0	0	0	0	0.05	8.0	0.5	300	0	-45
		26	98.0	1.5	0.5	0.03	0	0	0.24	4.2	5	700	-15	-25

Table 1 (Continued)

#1	Specimen No.	Composition Ratio					Electrical Characteristics						
		mol%			wt%		C ^{*2} ($\mu\text{F}/\text{cm}^2$)	$\tan\delta$ ^{*3} (%)	IR ^{*4} (M Ω) (10 ³)	Vb ^{*5} (V)	T.C. (%)		
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃					SiO ₂	-25°C	+85°C
	27	98.0	1.5	0.5	0.05	0	0	0.23	4.0	5	700	-10	-27
	28	98.0	1.5	0.5	0.10	0	0	0.20	4.0	5	700	0	-23
	29	98.0	1.5	0.5	0.20	0	0	0.20	4.0	5	700	+5	-20
	30	98.0	1.5	0.5	0.50	0	0	0.18	3.5	5	700	+5	-16
	31	98.0	1.5	0.5	0.80	0	0	0.14	3.5	5	1000	+6	-14
	32	98.0	1.5	0.5	1.00	0	0	0.04	3.0	5	1000	+5	-10
	33	98.0	1.5	0.5	0.20	0.003	0	0.15	4.0	7	900	+3	-20
	34	98.0	1.5	0.5	0.20	0.005	0	0.20	3.6	10	1100	+5	-20
	35	98.0	1.5	0.5	0.20	0.03	0	0.18	3.5	20	1500	+4	-20
	36	98.0	1.5	0.5	0.20	0.05	0	0.20	3.5	20	1500	+5	-20
	37	98.0	1.5	0.5	0.20	0.10	0	0.20	3.3	20	1500	+5	-20
	38	98.0	1.5	0.5	0.20	0.15	0	0.15	3.3	20	1300	+3	-20
	39	98.0	1.5	0.5	0.20	0.25	0	0.14	3.0	10	1000	+4	-18

X

As can be seen from Table 1, specimen Nos. 1, 2, 3, 10, 17, 23, 24, 25, 32, 40 and 44 are not included in the scope of the invention. More particularly, Table 1 indicates that Nb_2O_5 in an amount below 0.2 mol% or exceeding 3.0 mol% causes the capacitor to be decreased in unit area capacity C and increased in dielectric loss tan as well as decreased in insulation resistance IR and D.C. breakdown voltage Vb (Specimen Nos. 1, 2, 3, 23, 24 and 25). Also, it indicates that CeO_2 below 0.2 mol% causes the capacitor to be decreased in capacity and increased in dielectric loss; whereas when the amount of CeO_2 is above 3.0 mol%, the capacitor is increased in dielectric loss and exhibits improper temperature characteristics (Specimen Nos. 1, 3, 10, 17, 23 and 25). Co_3O_4 in an amount above 0.8 wt% and MnCO_3 above 0.25 wt% caused the capacity to be decreased (Specimen No. 32) and the D.C. breakdown voltage to be decreased (Specimen No. 40), respectively. Further, SiO_2 exceeding 0.25 wt% led to a decrease in capacity, insulation resistance and D.C. breakdown voltage of the capacitor (Specimen No. 44).

Thus, it will be noted that the semiconductive ceramic composition of the present invention effectively exhibits the above-noted advantages.

Example 2

Example 1 was substantially repeated except that BaCO_3 , TiO_2 , Nb_2O_5 , CeO_2 , Co_3O_4 , MnCO_3 , SiO_2 , SrCO_3 and Y_2O_3 in such amounts as shown in Table 2 were used as starting materials. The results were as shown in Table 2.

Table 2

*1 Specimen No.	Composition Ratio							Electrical Characteristics						
	mol%			wt%				C*2 ($\mu\text{F}/\text{cm}^2$)	tan δ *3 (%)	IR*4 (M Ω) (10 ³)	Vb*5 (V)	T.C.*6 (%)		
	BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃	SiO ₂	SrTiO ₃					Y ₂ O ₃	-25°C	+85°C
X 1	99.8	0.1	0.1	0	0	0	0	0	0.02	10.0	0.5	700	0	-10
X 2	99.4	0.1	0.5	0	0	0	0	0	0.05	7.0	0.7	500	+1	-13
X 3	95.9	0.1	4.0	0	0	0	0	0	0.10	8.0	0.7	500	+8	-30
4	99.6	0.2	0.2	0	0	0	0	0	0.15	4.3	5	800	+3	-10
5	99.3	0.2	0.5	0	0	0	0	0	0.18	4.0	5	700	+3	-15
6	96.8	0.2	3.0	0	0	0	0	0	0.25	4.3	7	1000	-1	-27
7	99.0	0.5	0.5	0	0	0	0	0	0.27	3.5	5	700	+4	-18
X 8	98.4	1.5	0.1	0	0	0	0	0	0.05	11.0	0.7	500	+5	-13
9	98.3	1.5	0.2	0	0	0	0	0	0.17	4.3	5	700	+5	-15
10	98.0	1.5	0.5	0	0	0	0	0	0.20	4.0	5	700	+5	-20
11	97.0	1.5	1.5	0	0	0	0	0	0.23	3.7	5	700	+1	-25
12	95.5	1.5	3.0	0	0	0	0	0	0.26	4.0	5	1000	-3	-30
X 13	94.5	1.5	4.0	0	0	0	0	0	0.28	7.0	5	1000	-10	-40

Table 2 (Continued)

#1	Specimen No.	Composition Ratio							Electrical Characteristics						
		mol%			wt%				C ^{*2} ($\mu\text{F}/\text{cm}^2$)	tan δ ^{*3} (%)	IR ^{*4} (M Ω) (10 ³)	V _b ^{*5} (V)	T.C. ^{*6} (%)		
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃	SiO ₂	SrTiO ₃					Y ₂ O ₃	-25°C	+85°C
	14	96.8	3.0	0.2	0	0	0	0	0	0.15	4.0	5	700	+5	-20
	15	96.5	3.0	0.5	0	0	0	0	0	0.15	4.5	5	500	+5	-23
	16	94.0	3.0	3.0	0	0	0	0	0	0.20	4.3	5	800	-2	-30
X	17	95.9	4.0	0.1	0	0	0	0	0	0.05	12.0	0.5	300	+13	-25
X	18	95.5	4.0	0.5	0	0	0	0	0	0.07	8.0	0.7	300	+7	-30
X	19	92.0	4.0	4.0	0	0	0	0	0	0.05	8.0	0.5	300	0	-45
	20	98.0	1.5	0.5	0.03	0	0	0	0	0.24	4.2	5	700	-15	-25
	21	98.0	1.5	0.5	0.10	0	0	0	0	0.20	4.0	5	700	0	-23
	22	98.0	1.5	0.5	0.20	0	0	0	0	0.20	4.0	5	700	+5	-20
	23	98.0	1.5	0.5	0.80	0	0	0	0	0.14	3.5	5	1000	+6	-14
X	24	98.0	1.5	0.5	1.00	0	0	0	0	0.04	3.0	5	1000	+5	-10
	25	98.0	1.5	0.5	0.20	0.05	0	0	0	0.20	3.5	20	1500	+5	-20
	26	98.0	1.5	0.5	0.20	0.25	0	0	0	0.14	3.0	10	1000	+4	-18

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Table 2 (Continued)

#1	Specimen No.	Composition Ratio					Electrical Characteristics								
		mol%		wt%			C ^{*2} ($\mu\text{F}/\text{cm}^2$)	tan δ ^{*3} (%)	IR ^{*4} (M Ω) (10 ³)	V _b ^{*5} (V)	T.C. ^{*6} (%)				
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃					SiO ₂	SrTiO ₃	Y ₂ O ₃	-25°C	+85°C
		98.0	1.5	0.5	0.20	0.35	0	0	0	0.18	2.0	5	300	+5	-15
		98.0	1.5	0.5	0.20	0.05	0.05	0	0	0.20	3.6	20	1500	+5	-20
		98.0	1.5	0.5	0.20	0.05	0.25	0	0	0.14	4.0	10	1000	+3	-18
X	30	98.0	1.5	0.5	0.20	0.05	0.35	0	0	0.05	5.0	1	200	+3	-15
X	31	98.0	1.5	0.5	0.20	0.05	0.05	0.05	0.2	0	3.4	20	1500	+5	-20
	32	98.0	1.5	0.5	0.20	0.05	0.05	0.05	0.5	0	2.5	20	1800	+5	-21
	33	98.0	1.5	0.5	0.20	0.05	0.05	0.05	5.0	0	2.5	10	1500	+8	-23
	34	98.0	1.5	0.5	0.20	0.05	0.05	0.05	10.0	0	2.3	10	1200	+10	-26
	35	98.0	1.5	0.5	0.20	0.05	0.05	0.05	20.0	0	2.2	5	850	+15	-30
X	36	98.0	1.5	0.5	0.20	0.05	0.05	0.05	30.0	0	2.0	0.2	350	-10	-65
X	37	98.0	1.5	0.5	0.20	0.05	0.05	0.05	20.0	0	2.2	5	850	+15	-30
X	38	98.0	1.5	0.5	0.20	0.05	0.05	0	0.05		3.5	20	1500	+5	-21
X	39	98.0	1.5	0.5	0.20	0.05	0.05	0	3.0		3.3	10	2000	+5	-20

Table 2 (Continued)

#1 Specimen	Composition Ratio							Electrical Characteristics							
	mol%			wt%				C*2 ($\mu\text{F}/\text{cm}^2$)	$\tan\delta$ *3 (%)	IR*4 (M Ω) (10^3)	Vb*5 (V)	T.C.*6 (%)			
	BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃	SiO ₂	SrTiO ₃					Y ₂ O ₃	-25°C	+85°C	
X 40	98.0	1.5	0.5	0.20	0.05	0.05	0.05	0.2	0.1	0.20	3.5	20	1800	+5	-20
41	98.0	1.5	0.5	0.20	0.05	0.05	0.05	0.5	0.1	0.20	2.5	20	2000	+5	-20
42	98.0	1.5	0.5	0.20	0.05	0.05	0.05	20.0	0.1	0.18	2.1	8	1400	+10	-26
X 43	98.0	1.5	0.5	0.20	0.05	0.05	0.05	30.0	0.1	0.37	2.2	0.7	500	-7	-60
X 44	98.0	1.5	0.5	0.20	0.05	0.05	0.05	0.2	3.0	0.18	3.5	20	1800	+5	-20
45	98.0	1.5	0.5	0.20	0.05	0.05	0.05	0.5	3.0	0.18	2.3	20	2000	+3	-21
46	98.0	1.5	0.5	0.20	0.05	0.05	0.05	20.0	3.0	0.15	2.1	10	1500	+12	-29
X 47	98.0	1.5	0.5	0.20	0.05	0.05	0.05	30.0	3.0	0.38	2.1	0.6	700	-5	-62
X 48	98.0	1.5	0.5	0.20	0.05	0.05	0.05	0.5	4.0	0.05	7.4	10	1600	+6	-15
X 49	98.0	1.5	0.5	0.20	0.05	0.05	0.05	30.0	4.0	0.29	4.2	0.5	600	-7	-58

*1: X indicates specimens which are not included in the scope of the present invention.

*2: C indicates unit area capacity

Table 3

#1	Composition Ratio								Electrical Characteristics						
	Specimen No.	mol%			wt%					C*2 ($\mu\text{F}/\text{cm}^2$)	tan δ *3 (%)	IR*4 (M Ω) (10 ³)	Vb*5 (V)	T.C.*6 (%)	
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃	SiO ₂	CaTiO ₃	Y ₂ O ₃					-25°C	+85°C
X	1	99.8	0.1	0.1	0	0	0	0	0	0.02	10.0	0.5	700	0	-10
X	2	99.4	0.1	0.5	0	0	0	0	0	0.05	7.0	0.7	500	+1	-13
X	3	95.9	0.1	4.0	0	0	0	0	0	0.10	8.0	0.7	500	+8	-30
	4	99.6	0.2	0.2	0	0	0	0	0	0.15	4.3	5	800	+3	-10
	5	99.3	0.2	0.5	0	0	0	0	0	0.18	4.0	5	700	+3	-15
	6	96.8	0.2	3.0	0	0	0	0	0	0.25	4.3	7	1000	-1	-27
	7	99.0	0.5	0.5	0	0	0	0	0	0.27	3.5	5	700	+4	-18
X	8	98.4	1.5	0.1	0	0	0	0	0	0.05	11.0	0.7	500	+5	-13
	9	98.3	1.5	0.2	0	0	0	0	0	0.17	4.3	5	700	+5	-15
10	10	98.0	1.5	0.5	0	0	0	0	0	0.20	4.0	5	700	+5	-20
11	11	97.0	1.5	1.5	0	0	0	0	0	0.23	3.7	5	700	+1	-25
12	12	95.5	1.5	3.0	0	0	0	0	0	0.26	4.0	5	1000	-3	-30
X	13	94.5	1.5	4.0	0	0	0	0	0	0.28	7.0	5	1000	-10	-40

Table 3 (Continued)

#1	Specimen No.	Composition Ratio					Electrical Characteristics								
		mol%		wt%			C*2 ($\mu\text{F}/\text{cm}^2$)	tan δ *3 (%)	IR*4 (M Ω) (10^3)	Vb*5 (V)	T.C.*6 (%)				
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	CO ₃ O ₄	MnCO ₃					SiO ₂	CaTiO ₃	Y ₂ O ₃	-25°C	+85°C
X	27	98.0	1.5	0.5	0.20	0.35	0	0	0	0.18	2.0	5	300	+5	-15
	28	98.0	1.5	0.5	0.20	0.05	0.05	0	0	0.20	3.6	20	1500	+5	-20
	29	98.0	1.5	0.5	0.20	0.05	0.25	0	0	0.14	4.0	10	1000	+3	-18
X	30	98.0	1.5	0.5	0.20	0.05	0.35	0	0	0.05	5.0	1	200	+3	-15
X	31	98.0	1.5	0.5	0.20	0.05	0.05	0.1	0	0.20	3.6	20	1500	+5	-20
	32	98.0	1.5	0.5	0.20	0.05	0.05	0.2	0	0.22	3.6	20	1900	+1	-17
	33	98.0	1.5	0.5	0.20	0.05	0.05	2.0	0	0.25	3.8	20	2000	-3	-15
	34	98.0	1.5	0.5	0.20	0.05	0.05	10.0	0	0.23	4.0	10	1800	-15	-5
	35	98.0	1.5	0.5	0.20	0.05	0.05	15.0	0	0.20	4.5	7	1500	-22	+3
X	36	98.0	1.5	0.5	0.20	0.05	0.05	20.0	0	0.05	8.0	5	1100	-27	+82
X	37	98.0	1.5	0.5	0.20	0.05	0.05	15.0	0	0.20	4.5	7	1500	-22	+3
X	38	98.0	1.5	0.5	0.20	0.05	0.05	0	0.05	0.20	3.5	20	1500	+5	-21
X	39	98.0	1.5	0.5	0.20	0.05	0.05	0	3.0	0.16	3.3	10	2000	+5	-20

Table 3 (Continued)

- *4: IR indicates insulation resistance.
*5: Vb indicates D.C. breakdown voltage
*6: T.C. indicates temperature characteristics.

Table 3 indicates that Nb_2O_5 in an amount below 0.2 mol% or exceeding 3.0 mol% causes the capacitor to be decreased in unit area capacity C and increased in dielectric loss $\tan \delta$, as well as decreased in insulation resistance IR and D.C. breakdown voltage Vb (Specimen Nos. 1, 2, 3, 17, 18 and 19). Also, it indicates that CeO_2 below 0.2 mol% causes the capacitor to be decreased in capacity and increased in dielectric loss; whereas when the amount of CeO_2 is above 3.0 mol%, the capacitor is increased in dielectric loss and exhibits improper temperature characteristics (Specimen Nos. 1, 3, 8, 13, 17 and 19). Co_3O_4 in an amount above 0.8 wt% and MnCO_3 above 0.25 wt% caused the capacity to be decreased (Specimen No. 24) and the D.C. breakdown voltage to be decreased (Specimen No. 27), respectively. Further, SiO_2 exceeding 0.25 wt% resulted in a decrease in capacity, insulation resistance and D.C. breakdown voltage of the capacitor (Specimen No. 30).

Further, CaTiO_3 less than 0.2 wt% failed in a significant increase in capacity and D.C. breakdown voltage of the capacitor, whereas when CaTiO_3 exceeds 15.0 wt%, the capacitor was decreased in capacity, increased in dielectric loss and exhibited improper temperature characteristics (Specimen Nos. 31, 36, 42 and 46).

Furthermore, Table 3 indicates that Y_2O_3 in an amount below 0.1 wt% fails in a significant increase of D.C. breakdown voltage of the capacitor and Y_2O_3 above 3.0 wt% decreases the capacity (Specimen Nos. 31, 37, 38, 39, 43, 47 and 48).

Thus, it will be noted that the semiconductive ceramic composition of the present invention effectively exhibits the above-noted advantages.

Example 4

Example 1 was substantially repeated except that BaCO_3 , TiO_2 , Nb_2O_5 , CeO_2 , Co_3O_4 , MnCO_3 , SiO_2 and Y_2O_3 in such amounts as shown in Table 4 were used as starting materials. The results were as shown in Table 4.

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Table 4 (Continued)

1	Specimen No.	Composition Ratio						Electrical Characteristics					
		mol%			wt%			C ^{*2} ($\mu\text{F}/\text{cm}^2$)	$\tan\delta$ ^{*3} (%)	IR ^{*4} (M Ω) (10^3)	Vb ^{*5} (V)	T.C. ^{*6} (%)	
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃	SiO ₂					-25°C	+85°C
	14	96.8	3.0	0.2	0	0	0	0.15	4.0	5	700	+5	-20
	15	96.5	3.0	0.5	0	0	0	0.15	4.5	5	500	+5	-23
	16	94.0	3.0	3.0	0	0	0	0.20	4.3	5	800	-2	-30
X	17	95.9	4.0	0.1	0	0	0	0.05	12.0	0.5	300	+13	-25
X	18	95.5	4.0	0.5	0	0	0	0.07	8.0	0.7	300	+7	-30
X	19	92.0	4.0	4.0	0	0	0	0.05	8.0	0.5	300	0	-45
	20	98.0	1.5	0.5	0.03	0	0	0.24	4.2	5	700	-15	-25
	21	98.0	1.5	0.5	0.10	0	0	0.20	4.0	5	700	0	-23
	22	98.0	1.5	0.5	0.20	0	0	0.20	4.0	5	700	+5	-20
	23	98.0	1.5	0.5	0.80	0	0	0.14	3.5	5	1000	+6	-14
X	24	98.0	1.5	0.5	1.00	0	0	0.04	3.0	5	1000	+5	-10
	25	98.0	1.5	0.5	0.20	0.05	0	0.20	3.5	20	1500	+5	-20
	26	98.0	1.5	0.5	0.20	0.25	0	0.14	3.0	10	1000	+4	-18

Table 4 (Continued)

#1 Specimen No.		Composition Ratio							Electrical Characteristics					
		mol%			wt%				C ^{*2} ($\mu\text{F}/\text{cm}^2$)	tan δ ^{*3} (%)	IR ^{*4} (M Ω) (10 ³)	V _b ^{*5} (V)	T.C. ^{*6} (%)	
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃	SiO ₂	Y ₂ O ₃					-25°C	+85°C
X	27	98.0	1.5	0.5	0.20	0.35	0	0	0.18	2.0	5	300	+5	-15
	28	98.0	1.5	0.5	0.20	0.05	0.05	0	0.20	3.6	20	1500	+5	-20
	29	98.0	1.5	0.5	0.20	0.05	0.25	0	0.14	4.0	10	1000	+3	-18
X	30	98.0	1.5	0.5	0.20	0.05	0.35	0	0.05	5.0	1	200	+3	-15
X	31	98.0	1.5	0.5	0.20	0.05	0.05	0.05	0.20	3.5	20	1500	+5	-21
	32	98.0	1.5	0.5	0.20	0.05	0.05	0.10	0.23	3.5	20	2000	+5	-20
	33	98.0	1.5	0.5	0.20	0.05	0.05	1.0	0.21	3.2	20	2000	+3	-23
	34	98.0	1.5	0.5	0.20	0.05	0.05	3.0	0.16	3.3	10	2000	+5	-20
X	35	98.0	1.5	0.5	0.20	0.05	0.05	4.0	0.03	8.2	10	2000	+10	-10

*1: X indicates specimens which are not included in the scope of the present invention.

*2: C indicates unit area capacity.

*3: $\tan \delta$ indicates dielectric loss.

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Table 4 (Continued)

*4:	IR indicates insulation resistance.
*5:	Vb indicates D.C. breakdown voltage
*6:	T.C. indicates temperature characteristics.

Table 4 indicates that Nb_2O_5 in an amount below 0.2 mol% or exceeding 3.0 mol% causes the capacitor to be decreased in unit area capacity C and increased in dielectric loss $\tan \delta$, as well as decreased in insulation resistance IR and D.C. breakdown voltage Vb (Specimen Nos. 1, 2, 3, 17, 18 and 19). Also, it indicates that CeO_2 below 0.2 mol% causes the capacitor to be decreased in capacity and increased in dielectric loss; whereas when the amount of CeO_2 is above 3.0 mol%, the capacitor is increased in dielectric loss and exhibits improper temperature characteristics (Specimen Nos. 1, 3, 8, 13, 17 and 19). Co_3O_4 in an amount above 0.8 wt% and MnCO_3 above 0.25 wt% caused the capacity to be decreased (Specimen No. 24) and the D.C. breakdown voltage to be decreased (Specimen No. 27). Further, SiO_2 exceeding 0.25 wt% led to a decrease in capacity, insulation resistance and D.C. breakdown voltage of the capacitor (Specimen No. 30).

Furthermore, Table 4 indicates that Y_2O_3 in an amount below 0.1 wt% fails in a significant increase of D.C. breakdown voltage of the capacitor and Y_2O_3 above 3.0 wt% decreases the capacity and increases the dielectric loss (Specimen Nos. 31 and 35).

Thus, it will be noted that the semiconductive ceramic composition of the present invention effectively exhibits the above-noted advantages.

While the invention has been described with a certain degree of particularity with reference to the examples, obvious modifications and variations are possible in the light of the above teachings. It is therefore to be understood that within the scope of the appended claims, the invention may be practiced otherwise than as specifically described.

Claims

1. A semiconductive ceramic composition comprising:

a base component comprising BaTiO_3 ; and
a minor component comprising Nb and Ce;

said Nb and Ce being present in amounts of 0.2 to 3.0 mol% on Nb_2O_5 and CeO_2 bases, respectively.

2. A semiconductive ceramic composition as defined in Claim 1 further comprising Co, said Co being present in an amount of 0.8 wt% or less on a Co_3O_4 basis.

3. A semiconductive ceramic composition as defined in Claim 2 further comprising Mn, said Mn being present in an amount of 0.25 wt% or less on a MnCO_3 basis.

4. A semiconductive ceramic composition as defined in Claim 3 further comprising SiO_2 , said SiO_2 being present in an amount of 0.25 wt% or less.

5. A semiconductive ceramic composition as defined in Claim 1 further comprising an additive component comprising Co, Mn, SiO_2 and SrTiO_3 , said Co and Mn being present in amounts of 0.8 wt% or less and 0.25 wt% or less on Co_3O_4 and MnCO_3 bases, respectively, and said SiO_2 and SrTiO_3 being present in amounts of 0.25 wt% or less and 0.5 to 20.0 wt%, respectively.

6. A semiconductive ceramic composition as defined in Claim 5 further comprising Y, said Y being present in an amount of 0.1 to 3.0 wt% on a Y_2O_3 basis.

7. A semiconductive ceramic composition as defined in Claim 1 further comprising an additive component comprising Co, Mn, SiO_2 and CaTiO_3 , said Co and Mn being present in amounts of 0.8 wt% or less and 0.25 wt% or less on Co_3O_4 and

Table 3 (Continued)

#1	Specimen	Composition Ratio					Electrical Characteristics								
		mol%		wt%			C*2 ($\mu\text{F}/\text{cm}^2$)	tan δ *3 (%)	IR*4 (M Ω) (10 ³)	Vb*5 (V)	T.C.*6 (%)				
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃					SiO ₂	CaTiO ₃	Y ₂ O ₃	-25°C	+85°C
	No.														
	40	98.0	1.5	0.5	0.20	0.05	0.05	0.2	0.1	0.22	3.5	20	2000	+3	-18
	41	98.0	1.5	0.5	0.20	0.05	0.05	15.0	0.1	0.22	3.7	10	1900	-15	+5
X	42	98.0	1.5	0.5	0.20	0.05	0.05	20.0	0.1	0.08	6.3	7	1600	-24	+91
X	43	98.0	1.5	0.5	0.20	0.05	0.05	0.1	3.0	0.16	3.3	10	1700	+5	-21
	44	98.0	1.5	0.5	0.20	0.05	0.05	0.2	3.0	0.20	3.3	20	2000	+3	-19
	45	98.0	1.5	0.5	0.20	0.05	0.05	15.0	3.0	0.20	4.4	10	1800	-20	+2
X	46	98.0	1.5	0.5	0.20	0.05	0.05	20.0	3.0	0.07	7.1	7	1400	-27	+95
X	47	98.0	1.5	0.5	0.20	0.05	0.05	0.2	4.0	0.06	5.2	10	2000	+5	-18
X	48	98.0	1.5	0.5	0.20	0.05	0.05	15.0	4.0	0.05	4.8	7	1700	-18	-1

*1: X indicates specimens which are not included in the scope of the invention.

*2: C indicates capacity per unit area

*3: $\tan\delta$ indicates dielectric loss.

Table 3 (Continued)

#1	Composition Ratio				Electrical Characteristics										
	Specimen No.	mol%		wt%			C ^{*2} ($\mu\text{F}/\text{cm}^2$)	tan δ ^{*3} (%)	IR ^{*4} (M Ω) (10 ³)	V _b ^{*5} (V)	T.C. ^{*6} (%)				
		BaTiO ₃	Nb ₂ O ₅	CeO ₂	Co ₃ O ₄	MnCO ₃					SiO ₂	CaTiO ₃	Y ₂ O ₃	-25°C	+85°C
	14	96.8	3.0	0.2	0	0	0	0	0	0.15	4.0	5	700	+5	-20
	15	96.5	3.0	0.5	0	0	0	0	0	0.15	4.5	5	500	+5	-23
	16	94.0	3.0	3.0	0	0	0	0	0	0.20	4.3	5	800	-2	-30
X	17	95.9	4.0	0.1	0	0	0	0	0	0.05	12.0	0.5	300	+13	-25
X	18	95.5	4.0	0.5	0	0	0	0	0	0.07	8.0	0.7	300	+7	-30
X	19	92.0	4.0	4.0	0	0	0	0	0	0.05	8.0	0.5	300	0	-45
	20	98.0	1.5	0.5	0.03	0	0	0	0	0.24	4.2	5	700	-15	-25
	21	98.0	1.5	0.5	0.10	0	0	0	0	0.20	4.0	5	700	0	-23
	22	98.0	1.5	0.5	0.20	0	0	0	0	0.20	4.0	5	700	+5	-20
	23	98.0	1.5	0.5	0.80	0	0	0	0	0.14	3.5	5	1000	+6	-14
X	24	98.0	1.5	0.5	1.00	0	0	0	0	0.04	3.0	5	1000	+5	-10
	25	98.0	1.5	0.5	0.20	0.05	0	0	0	0.20	3.5	20	1500	+5	-20
	26	98.0	1.5	0.5	0.20	0.25	0	0	0	0.14	3.0	10	1000	+4	-18

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Table 2 (Continued)

*3:	$\tan \delta$ indicates dielectric loss.
*4:	IR indicates insulation resistance.
*5:	Vb indicates D.C. breakdown voltage
*6:	T.C. indicates temperature characteristics.

Table 2 indicates that Nb_2O_5 in an amount below 0.2 mol% or exceeding 3.0 mol% causes the capacitor to be decreased in unit area capacity C and increased in dielectric loss $\tan \delta$, as well as decreased in insulation resistance IR and D.C. breakdown voltage Vb (Specimen Nos. 1, 2, 3, 17, 18 and 19). Also, it indicates that CeO_2 below 0.2 mol% causes the capacitor to be decreased in capacity and increased in dielectric loss; whereas when the amount of CeO_2 is above 3.0 mol%, the capacitor is increased in dielectric loss and exhibits improper temperature characteristics (Specimen Nos. 1, 3, 8, 13, 17 and 19). Co_3O_4 in an amount above 0.8 wt% and MnCO_3 above 0.25 wt% caused the capacity to be decreased (Specimen No. 24) and the D.C. breakdown voltage to be decreased (Specimen No. 27). Further, SiO_2 exceeding 0.25 wt% led to a decrease in capacity, insulation resistance and D.C. breakdown voltage of the capacitor (Specimen No. 30).

Further, SrTiO_3 less than 0.5 wt% failed in a significant decrease in dielectric loss of the capacitor, whereas when SrTiO_3 exceeds 20.0 wt%, the capacitor failed to exhibit good temperature characteristics and was decreased in insulation resistance and D.C. breakdown voltage (Specimen Nos. 31, 36, 38, 39, 40, 43, 44, 47 and 49).

Furthermore, Table 2 indicates that Y_2O_3 in an amount below 0.1 wt% decreases D.C. breakdown voltage of the capacitor and Y_2O_3 above 3.0 wt% decreases the capacity (Specimen Nos. 37 and 48).

Thus, it will be noted that the semiconductive ceramic composition of the present invention effectively exhibits the above-noted advantages.

Example 3

Example 1 was substantially repeated except that BaCO_3 , TiO_2 , Nb_2O_5 , CeO_2 , Co_3O_4 , MnCO_3 , SiO_2 , CaCO_3 and Y_2O_3 in such amounts as shown in Table 3 were used as starting materials. The results were as shown in Table 3.



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EUROPEAN SEARCH REPORT

Application number

DOCUMENTS CONSIDERED TO BE RELEVANT			EP 87111158.9
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
X	JP - A - 61-99 207 (ASAHI CHEMICAL IND KK) * Totality * --	1	C 04 B 35/46 H 01 B 1/08
X	JP - A - 57-208 003 (KYORITSU YOUNGYO GEN) * Totality * --	1,2	
X	JP - A - 57-128 903 (MATSUSHITA ELEC IND KK) * Totality * ----	1,3-9, 11-13	
The present search report has been drawn up for all claims			TECHNICAL FIELDS SEARCHED (Int. Cl. 4)
			C 04 B H 01 B
Place of search VIENNA		Date of completion of the search 18-11-1987	Examiner BECK
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>			

MnCO₃ bases, respectively, and said SiO₂ and CaTiO₃ being present in amounts of 0.25 wt% or less and 0.2 to 15.0 wt%, respectively.

8. A semiconductive ceramic composition as defined in Claim 7 further comprising Y, said Y being present in an amount of 0.1 to 3.0 wt% on a Y₂O₃ basis. 5

9. A semiconductive ceramic composition as defined in Claim 2 further comprising an additive component comprising Co, Mn, SiO₂ and Y, said Co, Mn and Y being present in amounts of 0.8 wt% or less, 0.25 wt% or less and 0.1 to 3.0 wt% on Co₃O₄, MnCO₃ and Y₂O₃ bases, respectively, and said SiO₂ being present in an amount of 0.25 wt% or less. 10 15

10. A semiconductive ceramic composition as defined in any one of Claims 1 to 9, wherein said composition is compacted and sintered to cause a surface layer thereof to be insulated.

11. A semiconductive ceramic composition comprising: 20

a base component comprising BaTiO₃;

a minor component comprising Nb and Ce, said Nb and Ce being present in amounts of 0.2 to 3.0 mol% on Nb₂O₃ and CeO₂ bases, respectively; and 25

an additive component comprising Co, Mn, SiO₂ and SrTiO₃, said Co and Mn being present in amounts of 0.8 wt% or less and 0.25 wt% or less on Co₃O₄ and MnCO₃ bases, respectively, and said SiO₂ and SrTiO₃ being present in amounts of 0.25 wt% or less and 0.5 to 20.0 wt%, respectively. 30

12. A semiconductive ceramic composition comprising:

a base component comprising BaTiO₃; 35

a minor component comprising Nb and Ce, said Nb and Ce being present in amounts of 0.2 to 3.0 mol% on Nb₂O₃ and CeO₂ bases, respectively; and

an additive component comprising Co, Mn, SiO₂ and CaTiO₃, said Co and Mn being present in amounts of 0.8 wt% or less and 0.25 wt% or less on Co₃O₄ and MnCO₃ bases, respectively, and said SiO₂ and CaTiO₃ being present in amounts of 0.25 wt% or less and 0.2 to 15.0 wt%, respectively. 40

13. A semiconductive ceramic composition comprising: 45

a base component comprising BaTiO₃;

a minor component comprising Nb and Ce, said Nb and Ce being present in amounts of 0.2 to 3.0 mol% on Nb₂O₃ and CeO₂ bases, respectively; and 50

an additive component comprising Co, Mn, SiO₂ and Y, said Co, Mn and Y being present in amounts of 0.8 wt% or less, 0.25 wt% or less and 0.1 to 3.0 wt% on Co₃O₄, MnCO₃ and Y₂O₃ bases, respectively, and said SiO₂ being present in an amount of 0.25 wt% or less. 55